CERTAIN HEALTH HAZARDS FROM FISSION PRODUCTS AND FALLOUT

BY

ROLF BJÖRNERSTEDT

UPPSALA 1959

ALMQVIST & WIKSELLS BOKTRYCKERI AB

CERTAIN HEALTH HAZARDS FROM FISSION PRODUCTS AND FALLOUT

INAUGURAL DISSERTATION

BY

ROLF BJÖRNERSTEDT
Fil. lic., Upplands nation

By due permission of the Philosophical Faculty
of the University of Uppsala, to be publicly discussed in the lecture hall
of the Institute of Physics on December 12, 1959, at 10 a.m.,
for the degree of Doctor of Philosophy.

UPPSALA 1959
ALMQVIST & WIKSELLS BOKTRYCKERI AB

CONTENTS

I. Löw, K. and Björnerstedt, R., Products of instantaneous fission of U^{235} with thermal neutrons. *Arkiv för Fysik 13*, 85 (1957).

II. Björnerstedt, R., Gamma radiation from nuclear weapons fallout. *Arkiv för Fysik 16*, 293 (1959).

III. Prawitz, J., Löw, K. and Björnerstedt, R., Gamma spectra of gross fission products from thermal reactors. ICPUAE, P/149 (Geneva 1958).

IV. Björnerstedt, R., Some GM-counter investigations. Research Institute of National Defence (1955). *Arkiv för Fysik 11*, 104 (1956), abstract.

V. Björnerstedt, R. and Löw, K., Fallout β-activity in precipitation. *Arkiv för Fysik 16*, 315 (1959).

VI. Björnerstedt, R., Theoretical calculation of the dose rate distribution from β-emitters in an infinite sandwich model. Bone and Radiostrontium, Almqvist and Wiksell, Stockholm (1958).

VII. Björnerstedt, R. and Engström, A. Maximum permissible body burden of strontium-90. *Science 129*, 327 (1959).

VIII. Björnerstedt, R. and Engström, A., Dosage implications from microscopic distribution. Radioisotopes in the Biosphere, University of Minnesota (in press).

IX. Björnerstedt, R. and Häggroth, S., A method for determination of the strontium distribution in human bone. Submitted to Nature (1959).

* – 596193

SUMMARY

The release of nuclear energy on a large scale in reactors and weapons tests has caused great concern as to the possible health hazards incurred. This concern may be said to have two reasons: the knowledge that radiation causes damage to living cells and the lack of precise knowledge as to the extent of this damage. As a result there exists at present a tremendous scientific effort directed towards increasing our knowledge of the health hazards from radioactive materials. It is the purpose here to briefly review only that small part of this effort that has been done at the Research Institute of National Defence (FOA) in Stockholm. The present thesis is best described against this more general background as it is an integral part thereof.

The work at FOA has had as its aim an evaluation of the hazards from nuclear weapons test fallout and the related effects to be expected in the case of nuclear warfare. A description of this work is given considering the origin, transport and effects of fallout.

Radioactive composition of fallout

Our first attempt to describe the radioactive composition of fallout was made in 1955 by Björnerstedt, Löw and Ulvönäs [1]. It was realized at the time that different fissionable nuclides would give rise to different yield distributions of the elements produced in fission, as will differences in neutron energy for one and the same nuclide. Yet the lack of reliable experimental yield values prompted the use of thermal neutron fission of U^{235} and Pu^{239} as giving a fairly adequate description of nuclear weapons fallout. When new data accumulated this work was repeated by Löw and Björnerstedt (I) in 1957. Still however only thermal neutron fission was considered. The first calculation published of the fission product composition and γ-spectra after fast neutron fission was made in 1959 by Björnerstedt (II).

In connection with the radioactive composition of fallout the work on reactor-produced fission products should be mentioned. This has been made by Prawitz and Rydberg [2] and contains a description by isotope and element of the fission products produced in a nuclear reactor. These data have been combined with values on γ-decay to evaluate the gamma spectrum of fission product mixtures (III). The authors are Prawitz, Löw and Björnerstedt. Experimental studies of γ-spectra from reactor-produced fission products have been reported by Löw [2 a].

The calculations of γ-spectra from fallout presupposes a knowledge of the decay characteristics of individual nuclides. In some cases, when this information has not been found in the literature, a special investigation was initiated. This has been the case with, e.g. La^{142}, the γ-lines of which [3] were determined by Ryde and Herrlander and Ba^{140} whose γ-lines were investigated by Setterwall, Löw and Edvarson [4].

The fallout may however contain other radioactive elements than the fission products, notably neutron-induced activities and fissionable material that remains unchanged after the explosion. This was briefly considered in ref. [1], but the original work is that of Dyrssen and Nyman [5] for salt water and Edvarson [6] for soil-induced activities.

It is important to remember that the fallout activity is generally bound to particles. The period of particle formation is between 1–30 seconds, depending upon the weapon yield, as this is the time required for the fire ball to cool off to about 1500°C. During this time interval, three important phenomena occur which vary with time. One is the variation in temperature, another the change in chemical composition due to decay of the radioactivity and a third process is the successive inclusion into the fireball of large masses of air, soil and/or water. There may consequently be differences between the radioactive composition of the total fallout and that of individual particles. Such differences are usually termed fractionation of fallout. Some experimental results on fractionation phenomena have been discussed recently by Edvarson, Löw and Sisefsky [7].

The transport of fallout

Nuclear weapons usually release an appreciable portion of the created radioactivity into the atmosphere. Although the fallout may be spread in several different ways, e.g. through currents in the oceans or with the ground water, the important process is that of atmospheric transport. To forecast this transport is difficult but over the years the possibilities have certainly become successively better. This is to a large extent due to the work on numerical forecasting using electronic computers that Rossby, Bolin and coworkers have carried out at the Meteorological Institute of Stockholm University. Numerous references from 1954 until the present will be found in Tellus. Another group at the Military Weather Bureau in Stockholm has made contributions along the same lines. All this work is of a more general interest and its usefulness is thus not restricted to considerations of nuclear weapons fallout. The same may be said about investigations carried out e.g. on the vertical temperature gradient, Waltersson [8], the height of the tropopause, Nordström [9], the vertical movements in the atmosphere, Söderberg [10] and the variability of winds, Eriksson [11]. And the difference is only gradual when one considers work that is directly

applicable to the transport of nuclear weapons fallout, like simulation of mushroom clouds with radar-reflecting materials, Nordström and Wickerts [12] or rainout and washout of particulate matter from the atmosphere, Lindblom [13].

The preceding argument has served the purpose of pointing out the close relation between studies of meteorology and radioactive fallout. This has been especially true with reference to the worldwide fallout. Measurements of this were started at FOA in 1952 by Björnerstedt. Samples of precipitation were collected, prepared and their β-activity measured for a time interval of some months. Part IV describes the work of Björnerstedt in developing the stable GM-counters used in these measurements and part V is a description by Björnerstedt and Löw of the results obtained from 1953 to 1957. During the first years [14] it was possible to date the activity of the samples by comparing their decay with the simple $t^{-1.2}$ law that applies approximately to the fission product decay. In this way an estimate was also obtained of the buildup of longlived activities, notably Sr^{90} and Cs^{137}. This simple method of analysis, however, became increasingly difficult to use when tests of megaton weapons started to inject radioactivity into the stratosphere, where it sometimes stayed for a couple of years and during the process of fallout was continually mixed with debris from subsequent weapons tests. It should perhaps be noted that the existence of this long stratosphere reservoir time was not known previously and it has lately been indicated [23] that there is a difference in the apparent reservoir time depending upon the latitude of fallout injection, tests in the northern latitudes giving times of the order of half a year. The decay law $I = I_0 \cdot t^{-1.2}$, did not apply too well from 1955 and onwards. For this reason the cumulative deposit has not been calculated since mid-1957.

In order to avoid some of the difficulties with mixed samples the technique of gamma spectrometry was introduced. This development was made possible by the construction of a multichannel pulse analyzer, Setterwall, Hansén [15–17]. The gamma spectrometry was thereafter used in a great number of investigations by Aler, Edvarson, Löw and several others e.g. ref. [18–23]. To replace calculations of the total fallout, measurements of the deposit in soil were introduced whereby studies of the uneven geographical distribution of fallout were made possible. In this way it has been found that the geographical distribution is approximately correlated to the amount of precipitation. The resulting differences are small at least over time periods of a year or longer. This is also apparent from an extension of the network for precipitation sampling. In addition new methods were developed for sampling of precipitation as well as air from the ground level and up to 12 km height, Edvarson [24–25]. This gave more direct means of studying the behaviour of fallout in relation to meteorological parameters and from this followed a theoretical attempt by Edvarson,

Prawitz and Almgren [26, 27] to describe the atmospheric transport of fallout. Through this work [14–27] it has become possible to give a fairly accurate description both of the total fallout and that of certain longlived isotopes over Sweden.

The work of Sisefsky, Edvarson and Löw [7, 28, 29] has made it possible to isolate individual fallout particles and thus effectively avoid that the sample measured is a mixture from several weapons tests. Through this development more detailed fallout analyses may be possible.

Fallout in the biosphere

In 1955 a research program was initiated often referred to as the SPAR-program (from the initials in soil-plant-animal relationship). The object of this program was a study of fallout behaviour in the biosphere.

The fallout distribution in soil has been measured by Löw, Edvarson, Andersson and Häggroth [20, 22, 30]. It was found that most of the activity rests in the top 5 cm of uncultivated soil. It is generally agreed that among all the fission products, the long-lived isotopes Sr^{90} and Cs^{137} are of particular importance in relation to the effects in the biosphere and man. The uptake by plants of Sr^{90} and Cs^{137} has been investigated by Fredriksson *et al.* [31–33] at the Agricultural Research Institute in Ultuna. As was to be expected, large differences were found for different species and different types of soil and soil treatment. This work suggests the effectiveness of various protectionary procedures that may be used against uptake of fallout radioactivity.

Åberg and Ekman of the Veterinary Institute in Stockholm have carried out work [33–36] on the problem of the uptake of different fission products by some animals. Related to this are the measurements of Edvarson, Häggroth and Löw of Sr^{90} and Cs^{137} in milk which indicates the existence of a strong seasonal variation [20, 30, 37].

In the final step, the content of Cs^{137} and Sr^{90} in man has to be analyzed. A whole body counter has been built in cooperation with Aktiebolaget Atomenergi, Stockholm, and used to measure a number of individuals for Cs^{137} [38, 39]. Only a few samples of human bone have however been determined for Sr^{90}.

Effects on man

The fallout causes external and internal irradiation of man. The external dose has been evaluated through the work of Jung [40–43], Aler and Herrlander [44], and Björnerstedt and Löw (I and II). The internal dose is extremely difficult to calculate because of the intricate way that the fallout passes from the explosion of a nuclear weapon to the inclusion of different nuclides in man.

If one however chooses a specific body burden as a starting point the calculation is to some extent facilitated.

The dose from Cs^{137} in the body is fairly easy to estimate once the body burden is known. Our first attempt at a calculation of the dose to bone from Sr^{90} was made by Björnerstedt (VI) as part of a more extensive work on bone by Engström (at Karolinska Institutet) and Björnerstedt, Clemedson, Nelson. One of the more important results of this later work was that the maximum permissible body burden of 1 μC Sr^{90}, as recommended at the time by ICRP, was found to be questionable when short term intake was considered. Tentatively a value of 0.1 μC was suggested under these conditions. The evidence behind this suggestion was certainly not complete at the time as the importance of a high dose to a small number of cells was unknown. It still is rather uncertain. The argument has however been discussed further in two papers by Björnerstedt and Engström (VII, VIII) and the conclusions seem at present to be that two extreme values of the maximum permissible body burden should be considered: 1 μC Sr^{90} for continuous intake and around 0.1 μC Sr^{90} for short term intake. This is to be compared with the present ICRP recommendation of 2 μC Sr^{90} for continuous conditions. With respect to the maximum body burden it may furthermore be shown that the most conservative approach may in some cases be to assume a threshold relationship between dose and damage. This is probably not so, however, for continuous intake of Sr^{90}. The present fallout situation resembles continuous intake conditions in an approximate way. These conditions are discussed in more detail in VIII.

With reference to the discussion of parts VI–VIII it is evident that a detailed knowledge of the Sr^{90} distribution inside bone is desirable. Part IX, Björnerstedt and Häggroth, outlines the principles of a method that may be used to this end in studies of the present Sr^{90} burden in humans. No definite results are available as yet.

In addition to the work described, mention should be made of related work of Nelson *et al.* [45–48] on the somatic effects of radiation and of Lüning *et al.* on the possible genetic consequences.

ACKNOWLEDGEMENTS

The present thesis is based on work carried out at the Research Institute of National Defence in cooperation with a number of other laboratories as mentioned previously. The author is indebted to essentially all those whose work has been reviewed here for suggestions and discussions.

Especially my sincere thanks are to be expressed to Dr B. Aler for his stimulating guidance of this work over many years and to Dr K. Edvarson and Mrs K. Löw for the invaluable advice and suggestions they have given.

Dr T. Magnusson as head of the laboratory and personal friend has always given my work his generous interest.

Professor A. Engström has over the years given me a number of "private lessons" in biology and physics. Without this stimulation it is doubtful if the work on strontium reported here, had ever been done.

Finally I would like to thank Mrs A. Olofsson for her efficient help in typing the manuscripts and all the other work connected therewith.

REFERENCES

1. Björnerstedt, R, Löw, K. and Ulvönäs, S., Products of simultaneous fissions. FOA (1955).
2. Composition of products formed by thermal neutron fission of U^{235}.
   I. Prawitz, J. and Rydberg, J., Compilation of nuclear data of the fission products. *Acta Chem. Scan. 12*, 369 (1958).
   II. Prawitz, J. and Rydberg, J., Chemical composition of stable and radioactive fission products. *Acta Chem. Scan. 12*, 377 (1958).
   III. Prawitz, J., Isotopic composition and atomic weight of the fission product elements. *Acta Chem. Scan. 13*, 163 (1959).

2 a. Aler, B., Björnerstedt, R., Löw, K., Ulvönäs, S., Jung, B, Edvarson, K. and Dyrssen, D., γ-doses from mixed fission products. *Arkiv för Fysik 11*, 101 (1956), abstract.
3. Ryde, H. and Herrlander, C. J., Gamma transitions in the decay of La^{142}. *Arkiv för Fysik* 13, 177 (1958).
4. Setterwall, N. H., Löw, K. and Edvarson, K., Communication to Nuclear Data Sheets (1958).
5. Dyrssen, D. and Nyman, P. O., Neutron induced activity in salt water from a nuclear explosion. *Acta Radiol. 43*, 421 (1955).
6. Edvarson, K., Calculation of neutron induced activities. FOA (1958).
7. Edvarson, K., Löw, K. and Sisefsky, J., Fractionation phenomena in nuclear weapons debris. FOA (1959), (in press).
8. Waltersson, E., Synoptisk-klimatologisk studie av de vertikala temperaturgradienterna över Bromma åren 1946–1955. MVC (1959).
9. Nordström, S.-B., Statistiska uppgifter beträffande tropopausen över Sverige. MVC (1959).
10. Söderberg, B. O., Vertikalrörelsefält erhållna med Wiin-Nielsens två-parametermodell. MVC (1959).
11. Eriksson, T., Höjdvindens variabilitet. FOA (1959).
12. Nordström, S.-B. and Wickerts, S., Simulerade atombombmoln med hjälp av radarreflekterande material. MVC (in preparation).
13. Lindblom, G., Radioaktivt utfall genom nederbörd. FOA (1959).
14. Björnerstedt, R., Edvarson, K. and Johansson, A., Det radioaktiva nedfallet i Stockholmstrakten under tiden 15.4.53 till 1.3. 1956. FOA (1956).
15. Setterwall, N. H., Pulsanalysator enligt Hutchinson och Scarrot. FOA (1956).
16. Setterwall, N. H. and Hansén, J., Detaljerad beskrivning av pulsanalysator enligt Hutchinson och Scarrot. FOA (1958).
17. Hansén, J., Read-out system for the Hutchinson-Scarrot analyzer. *Nuclear Instruments 3*, 271 (1958).

18. Aler, B., Björnerstedt, R., Edvarson, K. and Löw, K., Radioactive fallout from atomic weapons tests. FOA (1956).
19. Edvarson, K. and Löw, K., The radioactive fallout in Sweden up to 1.7. 1957. FOA (1957).
20. Löw K. and Edvarson, K., Cs^{137} in Swedish milk and soil, *Nature 183*, 921 (1959).
21. Lindblom, G., The radioactive fallout in Sweden 1957–1958. FOA (1959).
22. Löw, K. and Andersson, I. Ö., The fission products Zr^{95} and Nb^{95} in Swedish soils. FOA (1959).
23. Lindblom, G., Recent fallout measurements in Sweden. FOA (1959).
24. Edvarson, K., A method for monthly collection of radioactive fallout. FOA (1957).
25. Edvarson, K., A device for air-sampling in the upper atmosphere. FOA (1957).
26. Prawitz, J. and Almgren, B., Partiklars vertikala utbredning i atmosfären: Lösning av diffusionsekvation med bivillkor på analogimaskin. FOA (1959).
27. Edvarson, K., Partiklars vertikala utbredning i atmosfären: Atmosfärmodeller. FOA (in preparation).
28. Sisefsky, J., A method for determination of particle penetration depths in a filter. *Nature 182*, 1437 (1958).
29. Sisefsky, J., Method for photographic identification of microscopical radioactive particles. FOA (1959), *Brit. J. Appl. Phys.* (in press 1959).
30. Häggroth, S., Determination of Sr^{90} in dried milk, milk and soil. FOA (1959).
31. Fredriksson, L., Eriksson, B. and Rasmusson, B., Undersökningar över några kulturväxters upptagande av Sr^{90} ur jord vid olika Sr/Ca-förhållande i jorden. FOA (1957).
32. Fredriksson, L., Eriksson, B. and Rasmusson, B., Uptake of strontium and caesium by plants grown in soils of different texture and different calcium and potassium content. FOA (1957).
33. Studies on soil–plant–animal interrelationships with respect to fission products. ICPUAE P/177 (Geneva 1958).
    a. Fredriksson, L. and Eriksson, B., Plant uptake of Sr^{90} and Cs^{137} from soils.
    b. Rasmusson, B. and Gahne, B., Plant-animal interrelationships with respect to the absorption of Sr^{90}.
    c. Edvarson, K., Löw, K. and Fredriksson, L., Transport of long-lived fission products in Swedish soil.
34. Åberg, B. and Ekman, L., Progress report on the metabolism of fission products in ruminants. FOA (1957).
35. Åberg, B. and Ekman, L, Strålningsbiologiska undersökningar vid Kliniska Centrallaboratoriet, Veterinärhögskolan, 1.1.–31.12. 1958. FOA (1959).
36. Ekman, L. and Åberg, B., Metabolism and biological effects of yttrium in mammalia. A review. FOA (1959).
37. Löw, K., Cs^{137} in Swedish milk. Results up to June 1959. FOA (1959).
38. Löw, K., Preliminary measurements on body radioactivity. FOA (1957).
39. Andersson, I. Ö., Gamma radioactivity of Swedish people measured during May and June 1959. FOA (1959).
40. Jung, B., Energy distribution of the gamma-dose from mixed fission products from Pu^{239}. FOA (1955).
41. Jung, B., Beräkning av skyddsfaktorer för hus. FOA (1956).
42. Jung, B., The energy distribution of the gamma dose-rate at different distances from an infinite, plane, homogeneous source of mixed fission products from Pu^{239}. FOA (1956).

43. Jung, B., Gammadoshastigheten i olika hustyper orsakad av gammastrålande markbeläggning. FOA (1959).
44. Aler, B. and Herrlander, C. J., The computation of infinite plane 30-year doses from radioactive fallout. FOA (1957).
45. Nelson, A. and Holma, B., Location and duration of radioactive particles in mice. *Acta Radiol. 49*, 145 (1958).
46. Nelson, A. and Carlqvist, B., Svenska föreningens för medicinsk fysik och teknik årsmötesförhandlingar 1957. 7. Strålningsriskerna från Cs^{137}. *Svensk Läkartidning 55*, 608 (1958).
47. Nelson, A., Holmberg, B. and Wallgren, E., The transfer of Sr^{90} from mother to foetus in mice. *Radiation Research* (in press).
48. Nelson, A. and Carlqvist, B., Attempts to influence retention of radiostrontium. (to be published).